

Effect of the Orbital Level Difference in Doped Spin-1 Chains

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Doping of a two-orbital chain with mobile $S = 1/2$ Fermions and strong Hund's rule couplings stabilizing the $S = 1$ spins strongly depends on the presence of a level difference among these orbitals. By DMRG methods we find a finite spin gap upon doping and dominant pairing correlations without level-difference, whereas the presence of a level difference leads to dominant charge density wave (CDW) correlations with gapless spin-excitations. The string correlation function also shows qualitative differences between the two models.

KEYWORDS: spin-1 systems, orbital level-difference, Mott-Hubbard transition, superconductivity, string-order

Hole doping of strongly coupled spin-liquid ground-states is of great importance for the understanding of the Mott-Hubbard transition. A very special spin-liquid is the antiferromagnetic (AF) $S = 1$ Heisenberg (HB) chain, which shows a finite spin gap¹⁾ and a hidden topological order, the string-order parameter.²⁾ Only recently it has become possible to study experimentally the effect of doping in a spin-1 chain with mobile holes in the system $\text{Y}_{2-x}\text{Ca}_x\text{BaNiO}_5$.^{3,4)} In this material, the $S = 1$ spins are formed by a strong ferromagnetic (FM) Hund's coupling J_H between the two active orbitals of Ni^{2+} . Theoretically, a number of interesting questions arises by doping of a $S = 1$ chain with mobile holes. The competition between FM order induced by the double exchange mechanism⁵⁾ and AF order can result in completely different magnetic properties, the spin gap can immediately be destroyed upon doping or might persist, and finally which correlation function characterized by the single correlation exponent K_ρ dominates in the thermodynamic limit.

In a previous paper,⁶⁾ we have investigated the properties of a doped spin-1 chain with a level difference between the two electrons forming the $S = 1$ spin. Such a situation is likely in a system where the $S = 1$ spins are formed by two electrons in different orbitals of the same ion, as it is the case in $\text{Y}_{2-x}\text{Ca}_x\text{BaNiO}_5$.⁷⁻⁹⁾ Here we will concentrate on the effect of a level difference, by comparing the above case with a system with electrons in two equivalent levels. The latter situation is more likely in a ladder-system. A few years ago, this problem has been studied by Fujimoto and Kawakami in a weak coupling approach, where two ferromagnetically coupled Hubbard-chains have been investigated and compared to a Hubbard chain ferromagnetically coupled to a Heisenberg-chain.¹⁰⁾ In their analysis it is found that the model with two equivalent chains remains gapful, with a finite string-order parameter and dominant CDW correlations ($K_\rho = 1/2$). In contrast, the model with a level-difference becomes gapless upon doping and

shows two gapless spin-modes. Similar conclusions have been drawn by Nagaosa and Oshikawa in a semi-classical approach with two coupled Hubbard chains,¹¹⁾ and it is pointed out that for low hole doping, the system remains gapful for equivalent doping on the chains irrespective of FM or AF couplings on the rungs, whereas the system becomes gapless upon doping for different hole concentrations on both chains. However, since both of these studies are based on weak-coupling or mean-field approaches, it is not clear whether the results are valid in this one-dimensional, strongly correlated system and direct numerical evidence is needed. In this Letter we would like to investigate and compare doping of a spin-1 chain with and without level difference in an unperturbative manner free of approximations by the density matrix renormalization group method (DMRG).¹²⁾ This method is ideal for the study of groundstate energies and equal-time correlation functions, and we determine the correlation exponent K_ρ and the string order parameter. Additionally we also calculate the magnetic susceptibility at finite temperatures by the thermal DMRG (TDMRG) method.^{13,14)}

The first model we consider is a model with equal particle concentrations on both legs, and we call this the symmetric model in the following. The $S = 1$ spins are formed by two ferromagnetically coupled t - J chains with additional AF couplings between next-nearest neighbor sites on opposite legs. The individual terms which define the model are given by hopping along the chain i

$$H_{\text{kin}}^{(i)} = -t \sum_{j,\sigma} \mathcal{P} \left(c_{j,i,\sigma}^\dagger c_{j+1,i,\sigma} + H.c. \right) \mathcal{P},$$

where the projection operator \mathcal{P} prohibits doubly occupied sites, and $c_{j,i,\sigma}^\dagger$ is the particle creation operator on rung j and leg i with spin σ . Further the AF couplings $J > 0$ on the chain i between nearest neighbors and the diagonal couplings $J_d > 0$ are given by

$$H_{\text{af}}^{(i)} = J \sum_j \left(\mathbf{S}_{j,i} \mathbf{S}_{j+1,i} - \frac{1}{4} n_{j,i} n_{j+1,i} \right)$$

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$$H_{\text{diag}} = J_d \sum_{j,l} \left(\mathbf{S}_{j,1} \mathbf{S}_{l,2} - \frac{1}{4} n_{j,1} n_{l,2} \right) (\delta_{l,j-1} + \delta_{l,j+1}),$$

where the notation is standard, the indices are the same as above, and $\delta_{j,l}$ denotes Kronecker's delta function. The strong Hund's coupling $J_H < 0$ acts on the rungs

$$H_{\text{FM}} = J_H \sum_j \mathbf{S}_{j,1} \mathbf{S}_{j,2}.$$

Finally our Hamiltonian reads

$$H_{\text{sym}} = H_{\text{FM}} + H_{\text{diag}} + \sum_i \left(H_{\text{af}}^{(i)} + H_{\text{kin}}^{(i)} \right).$$

The second model contains a level difference between the two electrons forming the $S = 1$ spins and the lower band is localized. We will refer to this model as the asymmetric model in the rest, and it consists of a t - J chain ferromagnetically coupled to a $S = 1/2$ HB chain, with additional AF couplings between next-nearest neighbors on opposite legs. The Hamiltonian is defined as

$$H_{\text{sym}} = H_{\text{FM}} + H_{\text{diag}} + H_{\text{af}}^{(1)} + H_{\text{kin}}^{(1)}.$$

For both models we restrict ourselves to parameter values with $J = J_d$ and $|J_H| \gg t, J, J_d$, and if not otherwise mentioned we set $-J_H = 10t = 20J = 20J_d$. In the DMRG calculations we use systems sizes of up to 2×256 sites and up to 1300 states per system- and environment-block. In the TDMRG we keep 80 states per block and use finite Trotter-time steps of $\Delta\tau = 0.2t$.

We have reported on the asymmetric model in a previous paper,⁶⁾ and we briefly summarize the main results here. In that system, each mobile hole creates a small FM cloud by the double exchange mechanism (polaron). There is a very weak AF interaction among the polarons giving the lowest lying, gapless spin-excitations. However, the polarons are only a weak perturbation on top of the underlying spin-liquid of the Haldane chain, and there is a second energy scale in the spin-sector of the order of the Haldane gap of the undoped system. This energy scale shows up in exponentially decaying spin-correlations at short to intermediate distances and a finite string-order. The dominant correlation in the thermodynamic limit is given by $2k_F$ and $4k_F$ CDW order and the correlation exponent is $K_\rho \approx 0.51$.

In the following we will perform a similar analysis for the symmetric model and compare these findings with the asymmetric model. We start with the investigation of the spin gap. At half filling and $J_H \gg J, J_d$, both models can be mapped to the Haldane $S = 1$ chain with effective couplings $J = J_{\text{eff}}^{\text{sym}}$ in the symmetric case and $J_{\text{eff}}^{\text{as}} = 3/4J$ in the asymmetric case. By finite size scaling we have numerically determined the spin gap from $\Delta_s = \lim_{T \rightarrow 0} \Delta_s(L; N = Ln)$, where $\Delta_s(L; N = Ln) = \Delta_s(L; N) = E_0(L; N; S^z = 1) - E_0(L; N; S^z = 0)$ and $E_0(L; N; S^z)$ is the groundstate energy of the system with N particles on L sites and the total spin component along the z -direction is S^z . The results are in excellent agreement with the above mapping and we find $\Delta_{\text{spin}}^{\text{sym}} \approx 0.41(1)J_{\text{eff}}^{\text{sym}} = 0.205(5)t$ for the symmetric model and $\Delta_{\text{spin}}^{\text{as}} \approx 0.41(1)J_{\text{eff}}^{\text{as}} \approx 0.154(5)t$ for the asymmetric model. In agreement with refs.,^{10,11)} we find com-

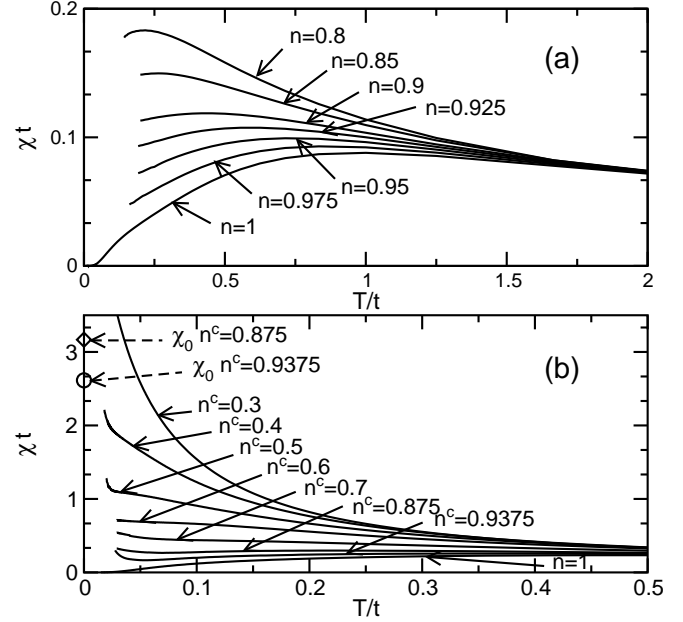


Fig. 1. Temperature dependencies of the magnetic susceptibility χ at different particle densities. Symmetric model (a) with $J = J_d = t/2$ and $J_H \rightarrow \infty$, and asymmetric model (b) with $-J_H = 10t = 20J = 20J_d$.

pletely different behavior for both models upon doping, as can be seen in Table I. While the symmetric model retains a rather large spin gap which decreases slightly upon doping (n is the particle density per single site), it is destroyed already for the smallest hole density of $n_h^c = 0.0625$ holes per site of the conduction band for the asymmetric model.

At finite temperatures near the gap value $T \propto J/2$, the magnetic susceptibility χ of both systems is strongly enhanced upon doping, and we show the TDMRG results in Fig. 1, where we have set $J_F \rightarrow \infty$ for the symmetric model for numerical reasons. This strong enhancement of χ is caused by the creation of a small FM cloud around each mobile hole by the double exchange mechanism, as the holes can gain kinetic energy by a FM alignment of the neighboring spins. At still lower temperatures, the magnetic susceptibility is suppressed for the symmetric model due to the spin gap and we find $\chi \rightarrow 0$ for $T \rightarrow 0$. In contrast, χ is finite and rather large at $T = 0$ for the asymmetric model, indicating the formation of larger FM moments due to the double exchange mechanism (see ref. 6). However, the groundstate is spin-singlet for both

Table I. Finite size scaling of the spin gap Δ_{spin} for $-J_H = 10t = 20J = 20J_d$ and different values of the hole density $n_h = 1 - n$ for the symmetric model and hole density in the conduction band n_h^c for the asymmetric model. Also listed is the spin-spin correlation length ξ .

n_h	$\Delta_{\text{spin}}^{\text{sym}}$	ξ^{sym}	$\Delta_{\text{spin}}^{\text{as}}$	ξ^{as}
0	0.205(5) t	6.05(5)	0.1504(5) t	6.05(5)
0.0625	0.148(5) t	6.1(2)	0	7.9(2)
0.125	0.120(5) t	6.7(2)	0	11.2(2)

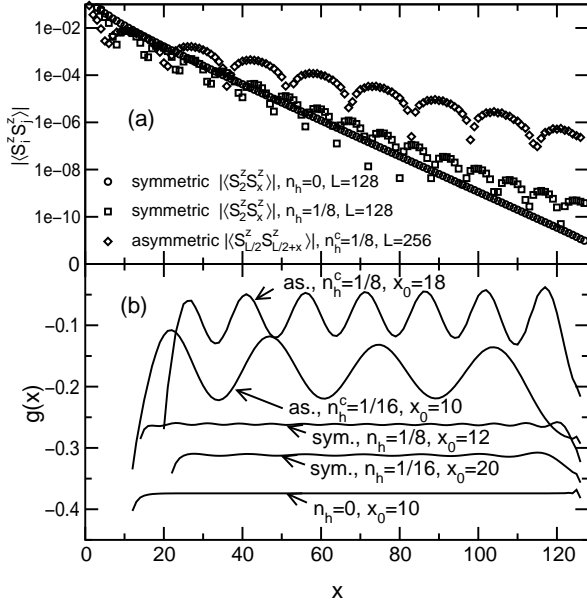


Fig. 2. Spin-spin correlations (a) for the symmetric and asymmetric model with $-J_H = 10t = 20J = 20J_d$, and string-order $g(x)$ for both models with same parameters (b).

models, as we have tested with the DMRG method by the calculation of $\sigma(i, j) = S_i^z S_j^z - 1/2(S_i^+ S_j^-)$ which vanishes for a rotationally invariant groundstate and the result is zero within the numerical precision of the DMRG for this quantity for both models ($|\sigma(i, j)| < 10^{-6} \forall i, j$).

Complementary information about the magnetic properties is provided by the spin-spin correlations shown in Fig. 2(a). At half filling we find exponentially decaying correlations as expected for gapful systems and the correlation length is $\xi \approx 6.03$.¹⁵⁾ Also at finite doping, the spin-spin correlations seem to decay exponentially for both systems. We have determined ξ by fitting to $S_i^z S_{i+x}^z \propto \cos(2k_F x) e^{-x/\xi}$ and the results are listed in Table I. In the case of the symmetric model, the correlation length increases only slightly upon doping to $\xi^{\text{sym}} \approx 6.7 \pm 0.2$ for $n_h = 1/8$, while the increase to $\xi^{\text{as}} \approx 11.2$ is considerably larger for the asymmetric model at the same density. In fact we expect a crossover to power-law decay at large distances in the latter case (see ref. 6). More insight about the spin-configuration can be gained from the string-order parameter $g(x) = \langle (\sum_{i=1,2} S_{x_0,i}^z) \left(\prod_{k=x_0+1, j=1,2}^{x-1} e^{i\pi S_{k,j}^z} \right) (\sum_{l=1,2} S_{x,l}^z) \rangle$, which reveals the hidden $Z_2 \times Z_2$ symmetry of the Haldane $S = 1$ chain and quickly approaches the value of $g(x) \approx -0.374$ for $x \gg 1$ in the undoped case.²⁾ From Fig. 2(b) it can be seen that $|g(x)|$ is reduced upon doping but remains finite for both models. A finite string order parameter for the symmetric model has also been obtained in ref. 10. Compared to the symmetric model, the reduction of $|g(x)|$ upon doping is roughly three times larger for the asymmetric model. In addition, the oscillations in $|g(x)|$ are a strong amplification of the corresponding Friedel oscillations in the charge density induced by the open boundary conditions. In fact, for the asymmetric model, we again expect a crossover of $|g(x)|$

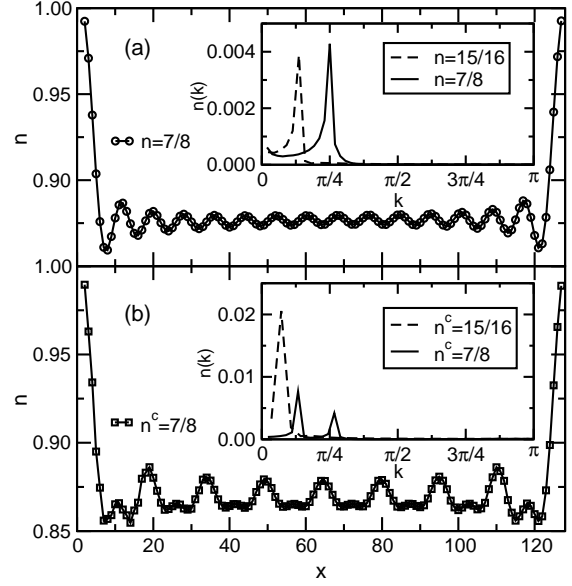


Fig. 3. Charge density for the symmetric model (a) at $n = 7/8$ and the symmetric model (b) at $n^c = 7/8$, the parameters for both models are $-J_H = 10t = 20J = 20J_d$ and the length is $L = 128$. In the insets we show the corresponding Fourier transforms $n(k)$.

to power-law decay at large distances after a sufficient number of oscillations. By comparison, $|g(x)|$ is constant within 1.2% for the symmetric model apart from boundary effects, and we conclude that its spin structure closely resembles that of the undoped case.

Let us turn to the charge sector next and start with the above mentioned Friedel oscillations in the charge density. In their Fourier transforms we only find one peak at $k = 2n\pi$ for the symmetric model. This corresponds to the $2k_F$ CDW fluctuations, since the lower band is fully occupied and hence there are $2(1-n)$ holes in the upper band, giving $2k_F = (1 + 1 - 2(1-n))\pi \equiv 2n\pi$ for a large Fermi volume. In the case of the asymmetric model we also find only one peak at $k = n_h^c \pi$ for the smallest hole densities $n_h^c = 1/16$, and two peaks at $k = n_h^c \pi$ and $k = 2n_h^c \pi$ for the larger hole doping $n_h^c = 1/8$. This is consistent with $2k_F$ and $4k_F$ CDW fluctuations for a large Fermi volume including the electrons of the lower orbital with $2k_F = (n^c + 1)\pi$ in that case. Having identified these $2k_F$ and $4k_F$ fluctuations, we can determine the correlation exponent K_ρ by fitting to the Friedel oscillations of an impurity potential. In the gapful case they are given by $\delta_n(x) \propto C_1 \cos(2k_F x) x^{-K_\rho} + C_2 \cos(4k_F x) x^{-2K_\rho}$ and $\delta_n(x) \propto C_1 \cos(2k_F x) x^{-(1+K_\rho)/2} + C_2 \cos(4k_F x) x^{-2K_\rho}$ in the gapless case.¹⁶⁾ The correlation exponents thus obtained are $K_\rho \approx 1.5 \pm 0.05$ for the symmetric case at $n_h = 7/8$ indicating dominant superconducting pairing correlations, and $K_\rho \approx 0.51 \pm 0.05$ for to asymmetric model at $n_h^c = 1/8$, giving dominant CDW correlations. For both cases, the sites near the boundaries need to be discarded for the fit because of trapped states, and the uncertainty stems from the fitting ambiguity. In the symmetric model hole pairs are formed on the rungs in order to gain the strong Hund's rule coupling J_F . The pair binding energy $\Delta_{\text{pair}} \approx 2.29t$ in the low-doping region

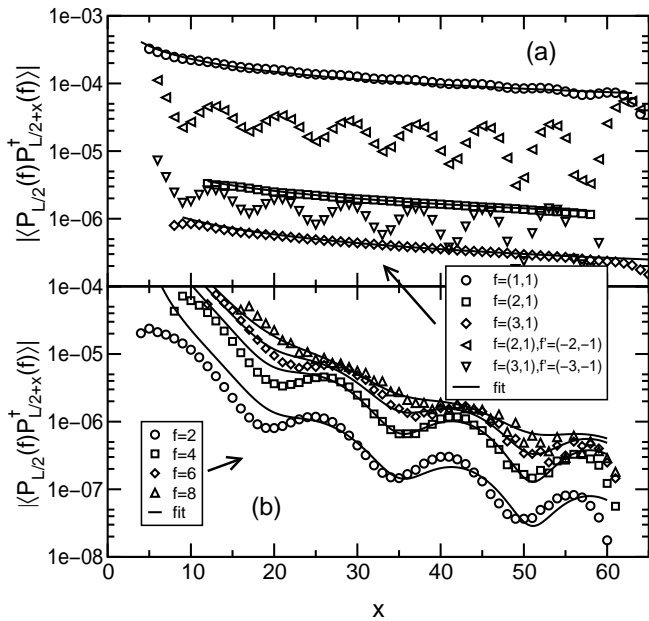


Fig. 4. Singlet pairing correlations $P_i(\mathbf{f})P_{i+x}^\dagger(\mathbf{f}')$ with $P_i^\dagger(\mathbf{f}) = \frac{1}{\sqrt{2}}(c_{i,\uparrow}^\dagger c_{i+f,\downarrow}^\dagger - c_{i,\downarrow}^\dagger c_{i+f,\uparrow}^\dagger)$ for (a) symmetric model at $n = 7/8$ and (b) asymmetric model at $n^c = 7/8$ and $-J_H = 10t = 20J = 20J_d$ for both models.

obtained from $\Delta_{\text{pair}} = 2E_1 - E_0 - E_2$ is correspondingly large, where E_n is the groundstate energy with n holes. This pair-binding mechanism does of course not work for the asymmetric model, but we still find a positive pair-binding energy $\Delta_{\text{pair}} \approx 0.016t$ in that case. Pair formation in that context has also been found in ref. 9. From a second structure that develops in the hole-pockets at larger doping, we expect that these pairs get spatially more extended upon doping.

The pairing correlations defined as $P_i(\mathbf{f}')P_{i+x}^\dagger(\mathbf{f})$ with $P_i^\dagger(\mathbf{f}) = \frac{1}{\sqrt{2}}(c_{i,\uparrow}^\dagger c_{i+f,\downarrow}^\dagger \mp c_{i,\downarrow}^\dagger c_{i+f,\uparrow}^\dagger)$ allow an independent calculation of the correlation exponent K_ρ , and provide further information on the form-factor \mathbf{f} of the pairs. From the numerous possibilities of the form-factor \mathbf{f} for the symmetric model, we restrict the discussion to those with the largest amplitudes in the following. At very short distances $d < 5$ triplet pairing correlations for rung pairs with $\mathbf{f} = (0, 1)$ have the largest amplitudes, but as the triplet correlation functions decay exponentially, the amplitudes for singlet pairs formed on the rungs with an extension of the pair over two to three rungs become larger at distances $d > 5$. We show the results for the singlet pairs with the largest amplitudes in Fig. 4. These correlations decay as $P_i(\mathbf{f})P_{i+x}^\dagger(\mathbf{f}) \propto x^{-1/K_\rho}$ for a gapful Luther-Emery liquid,¹⁷⁾ and surprisingly there are almost no $2k_F$ fluctuations for the largest pairing correlations despite the Friedel oscillations in the charge density. The correlation exponent determined from the fit $K_\rho \approx 1.55 \pm 0.05$ is in excellent agreement with the result obtained from the charge-density. For comparison, the correlation exponent obtained by weak-coupling is $K_\rho = 0.5$.¹⁰⁾ The form factor of the pairs with largest amplitudes $\mathbf{f} = (1, 1), (2, 1)$ and $(3, 1)$ are consistent with a d_{x-y} symmetry analogue for a ladder

with vanishing $\mathbf{f} = (0, 1)$ and $(1, 0)$ amplitudes. For the asymmetric model we obtain $K_\rho \approx 0.51 \pm 0.05$ for a gapless Tomonaga-Luttinger liquid with $P_i(\mathbf{f})P_{i+x}^\dagger(\mathbf{f}) \propto A_0 \ln(x)^{-1.5} x^{-1-1/K_\rho} + A_2 \cos(2k_F x) x^{-K_\rho-1/K_\rho}$ ¹⁸⁾ by simultaneously fitting to $f = 2, 4, 6$, and 8 , again in excellent agreement with the previous fit.

In conclusion we have studied the effect of a level difference on doping of two-orbital chains. In addition to a finite spin gap for the symmetric model and gapless excitations for the asymmetric model also obtained by weak-coupling theories,^{10, 11)} we find a new difference in the dominant correlation functions, which consist of pairing correlations with $K_\rho \approx 1.5$ for the symmetric model and CDW correlations with $K_\rho \approx 0.5$ for the asymmetric model. Further the string-order parameter remains finite for the symmetric model, however strongly reduced by the polarons in the asymmetric model.

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